

Amendments to the Specification:

Please replace the paragraphs on page 11, line 24 – page 12, line 19, with the following amended paragraphs:

In contrast, a compound which should not be included in the inhibitor of (a) means a compound which does not have the effect of preventing Cu from corroding or which does not have a Cu-trapping effect at all. If a compound has such an effect even to a little extent, the compound is included in the present invention.

As an organic inhibitor which can be used, ~~the one~~ ones where a central element of a polar group of said organic inhibitor contains at least one selected from the group consisting of N, P and As in 5B group and O, S and Se in 6B group of ~~a~~ the periodic table is preferable:

As the aforementioned organic inhibitor, there may be preferably used one containing at least one of 1, 2, 3 – benzotriazole, 4 or 5 – benzotriazole, benzimidazole, 2 – benzimidazolethiol, 2 – benzoxazolethiole, 2 – ~~methylbensothiazole~~, methylbenzothiazole, indole, and 2-mercaptothiazoline.

As the aforementioned organic inhibitor, there may be preferably used ~~the one~~ containing dithiocarbamic acid or a derivative thereof. As the derivative, there is preferably used one containing at least one of diethyldithiocarbamate, dimethyldithiocarbamate, N – methyldithiocarbamate, ethylene-bisdithiocarbamate, and dithiocarbamate.

Please replace the paragraph on page 13, lines 9-12, with the following amended paragraph:

As the aforementioned¹ inhibitor, there is preferably used one containing at least one of amine type organic compound, ~~amid~~ amide type organic compound, tetrazole derivative, 3 – amino type organic compound, and 1, 2, 4 – triazole type organic compound.

Please replace the paragraph on page 13, line 19 – page 14, line 10, with the following amended paragraph:

These organic inhibitors are suitably used because they are stable in an electrolyte solution and have high Li^+ conductivity. Content of these organic inhibitors in a nonaqueous electrolyte solution is preferably within 0.01 – 10.0 mass%, and more preferably 0.10 – 0.50 mass%. As is clear from Examples described below, when the content of the organic inhibitors in a nonaqueous electrolyte solution is 0.01 mass%, effect as a Cu-corrosion inhibitor or a Cu-trapping agent is a little, and therefore when it is used in a battery, the battery has little functional effect. On the other hand, when the content of the organic inhibitors in a nonaqueous electrolyte solution is 10.0 mass% or more, the battery characteristics deteriorate in total as battery reaction conversely ~~though through~~ effect as a Cu-corrosion inhibitor or a Cu-trapping agent increases. The reason why the battery characteristics deteriorate is not clear but seems to be due to decrease in ion conductivity because an electrolyte solution is diluted if the content of inhibitors is too much.

Please replace the paragraphs on page 15, line 13 – page 16, line 8, with the following amended paragraphs:

These inorganic inhibitors are suitably used because they are stable in an electrolyte solution and show high Li^+ conductivity. Content of these inorganic inhibitors in a nonaqueous electrolyte solution is preferably 0.01 – ~~0.10~~ 10.0 mass% and more preferably 0.10 – 0.50 mass%. When the content of the inorganic inhibitor in a nonaqueous electrolyte solution is 0.01 mass%, effect as a Cu-corrosion inhibitor or a Cu-trapping agent is a little, and therefore when it is used in a battery, the battery has little functional effect. On the other hand, when the content of the inorganic inhibitor in a nonaqueous electrolyte solution is 10.0 mass% or more, properties of the battery deteriorate in total as battery reaction conversely ~~though through~~ effect

as a Cu-corrosion inhibitor or a Cu-trapping agent increases. Like the case of organic inhibitor, the reason why the battery characteristics deteriorate is not clear but seems to be due to decrease in ion conductivity because an electrolyte solution is diluted if the content of inhibitors is too much.

Here, the mechanism of suppressing corrosion of Cu by an inorganic inhibitor and trapping mechanism of the present invention is described.

Please replace the paragraph on page 16, lines 19-25, with the following amended paragraph:

Further, if the compound is an organic compound having a heteroatom, HF in an electrolyte solution can be trapped by the effect of the heteroatom. Though, as matter of course, corrosion of a battery and deterioration of a nonaqueous electrolyte solution can be suppressed, hindrance to electric reaction can be remarkably reduced ~~synergically~~ synergistically because elution of Cu in an electrolyte solution is suppressed.

Please replace the paragraphs on page 19, line 23 – page 20, line 19, with the following amended paragraphs:

Therefore, since HF can be immobilized to be in inactive condition because an electron-donating element in a portion of an organic base of the compound and a substituent show Lewis basicity, ~~and thereby~~ the reaction between HF and battery members is suppressed. In addition, before the aforementioned SEI composite is formed, an anion of inorganic acid of the compound reacts with Li^+ to form a salt (LiCl , Li_2SO_4) and cover a surface of a negative active material. The film covering the surface of the negative active material is of a salt of a strong acid, which is chemically stable. This enables ~~to suppress~~ suppression of direct contact between the negative active material and HF and ~~to suppress~~ suppression of further growth of a SEI layer.

By the way, in the present invention, the SEI layer of a strong acid salt, which is intentionally generated on a surface of the negative active material, does not hinder movement of Li^+ to a gap between negative-electrode carbon layers. This is because the quantity of strong acid anions, which becomes a material of a strong acid salt, in an electrolyte can be controlled depending on ~~a~~ the quantity of the compound added. This enables ~~to form~~ formation of a SEI layer on a surface of a negative active material by controlling the quantity of anions in a range where movement of Li^+ is not hindered and where a conventional SEI layer cannot be formed.

Please replace the paragraphs on page 21, line 14 – page 22, line 4, with the following amended paragraphs:

Examples are 2, 2, 6, 6 – tetramethyl – 1 – piperidinyloxy free radical, 4 –cyano –2, 2, 6, 6 – tetramethyl – 1 – piperidinyloxy free radical, and 3 – cyano – 2, 2, 5, 5 – tetramethyl – 1 – pyrrolidinyloxy free radical. These are small in molecular skeleton, quickly ~~reacts~~ react with a radical molecule generated from an organic solvent, are stable in an electrolyte solution, and do not hinder movement of Li^+ in the electrolyte solution; and thereby ~~being~~ they are suitably used as the compound.

Next, the aforementioned compound (d) is described.

As a cyclic compound which becomes a Mn^{2+} supplier, manganese (II) phthalocyanine or ~~a~~ manganese (II) phthalocyanine derivatives ~~is~~ are suitably used.

Specifically, manganese (II) phthalocyanine shown by the following chemical formula (IX) exemplifies the compound. This is stable in an electrolyte solution and exhibits high Li^+ conductivity, and therefore ~~being suitable~~ is suitably used as the compound.

Please replace the paragraph on page 23, lines 8-12, with the following amended paragraph:

As a method for controlling a radical decomposition reaction as ~~the~~ above, two methods may be employed: 1) adding a radical compound and 2) utilizing chemical equilibrium reaction for extinguishing the radical shown by the following formulae 2) and 3), respectively.

Please replace the paragraph on page 24, line 18 – page 25, line 1, with the following amended paragraph:

HF generated at this time ~~elute~~elutes transition metal by dissolving a positive active material with dissolving and corroding metal material of a battery case and a current collector to derive formation of vicious SEI containing a metallic atom. Incidentally, decomposition of an electrolyte is accelerated and proceeded more with a temperature of a battery being higher. Since the SEI formation reaction is an exothermic reaction, decomposition of an electrolyte by water is accelerated by this heat, and HF is further formed.

Please replace the paragraphs on page 26, line 6 – page 27, line 2, with the following amended paragraphs:

HF generated at this time ~~elute~~elutes transition metal by dissolving a positive active material with dissolving and corroding metal material of a battery case and a current collector to drive formation of vicious SEI containing a metallic atom. Incidentally, decomposition of an electrolyte is accelerated and proceeded more with a temperature of a battery being higher. Since the SEI formation reaction is an exothermic reaction, decomposition of an electrolyte by water is accelerated by this heat, and HF is further formed.

Therefore, an atom showing Lewis acidity, that is, an Al atom having an empty electron orbit and showing an electron-attracting property is coordinately ~~bond~~bonded with a H₂O molecule present in the same electrolyte solution and having an unshared electron pair to fix H₂O in a molecular structure of the compound. In the same manner, an atom showing Lewis basicity, that is, a N atom having an unshared electron pair and showing an electron-donating property is coordinately bonded with HF having an empty electron orbit to fix HF in a molecular structure of the compound. By this, since the compound fixes HF even in the case that temperature of a battery itself becomes high while charge-discharge is repeated, formation of vicious SEI is suppressed.

Please replace the paragraph on page 27, line 13 – page 28, line 21, with the following amended paragraph:

Suitable three-dimensional siloxane compounds are specifically 1 – allyl – 3, 5, 7, 9, 11, 13, 15 – heptacyclopentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane, 1 – (3 – chloropropyl) – 3, 5, 7, 9, 11, 13, 15 – heptacyclo-pentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane, 1 – (4-vinylphenyl) – 3, 5, 7, 9, 11, 13, 15 - heptacyclopentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane, ethyl - 3, 5, 7, 9, 11, 13, 15 - heptacyclopentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane – 1 – undecanoate, 1, 3, 5, 7, 9, 11, 14 – heptacyclohexytricyclo [7. 3. 3. 1^{5,11}] heptasiloxane – 3, 7, 14 – triol, 1, 3, 5, 7, 9 11, 13 – heptacyclopentyl – 15 – [2 – (diphenylphosphino) ethyl] pentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane, 1, 3, 5, 7, 9, 11, 13 – ~~heptacyclopentyl~~heptacyclopentyl – 15 – glycidilpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane, 3, 5, 7, 9, 11, 13, 15 – heptacyclopentylpentacyclo {[9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane – 1 – butylonitrile, 3, 5, 7, 9, 11, 13, 15 – heptacyclopentylpentacyclo {[9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane – 1 – ole, 3 – (3, 5, 7, 9, 11, 13, 15 – ~~heptacyclopentyl~~pentacyclo

~~heptacyclopentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane – 1 – yl) propylmethacrylate,~~
 1, 3, 5, 7, 9, 11, 14 – heptacyclopentyltricyclo [7. 3. 3. 1^{5,11}] heptasiloxane – endo – 3, 7, 14 –
 triol, 1, 3, 5, 7, 9, 11, 13 – ~~heptacyclopenthy~~~~l~~~~heptacyclopentyl~~ – 15 – vinylpentacyclo [9. 5. 1.
 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane, 1 – hydride – 3, 5, 7, 9, 11, 13, 15 – ~~heptacyclopenthy~~~~l~~~~pentacyclo~~
~~heptacyclopentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}] octasiloxane, methyl – 3, 5, 7, 9, 11, 13, 15~~
~~– heptacyclopenthy~~~~l~~~~pentacyclo~~~~heptacyclopentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}]~~
 octasiloxane – 1 – propionate, 1 – [2-(5-norbornane – 2 – yl) ethyl] – 3, 5, 7, 9, 11, 13, 15 –
~~heptacyclopenthy~~~~l~~~~pentacyclo~~~~heptacyclopentylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}. 1^{7,13}]~~
 octasiloxane, 1, 3, 5, 7, 9, 11, 13, 15 – octakis (dimethylsilyloxy) pentacyclo [9. 5. 1. 1^{3,9}.
 1^{5,15}. 1^{7,13}] octasiloxane, and 1, 3, 5, 7, 9, 11, 13, 15 – octavinylpentacyclo [9. 5. 1. 1^{3,9}. 1^{5,15}.
 1^{7,13}] octasiloxane. Since this has a cyclic structure, it is stable in an electrolyte and has high
 Li⁺ conductivity; and thereby ~~being is~~ preferably used as the compound.

Please replace the paragraphs on page 28, line 24 – page 29, line 12, with the following amended paragraphs:

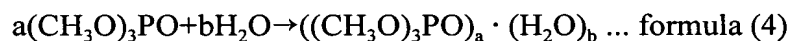
Water is present in an electrolyte solution of the present invention though the amount is very small; and an electrolyte solution is decomposed, and an electrolyte solution and an electrolyte are decomposed by the water to generate HF, gas (~~CO₂~~), etc.

HF generated at this time ~~elute~~elutes transition metal by dissolving a positive active material with dissolving and corroding metal material of a battery case and a current collector to drive formation of vicious SEI containing a metallic atom. Incidentally, decomposition of an electrolyte is accelerated and proceeded more with a temperature of a battery being higher. Since the SEI formation reaction is an exothermic reaction, decomposition of an electrolyte by water is accelerated by this heat, and HF is further formed.

Please replace the paragraphs on page 38, line 13 – page 39, line 23, with the following amended paragraphs:

As such a water-extracting agent, it is preferable to use a liquid agent which uniformly mixes with an electrolyte solution and with which the inside of an interior electrode body is impregnated uniformly. Water-extracting agents capable of being used in the present invention are specifically organic ~~phosphorous~~phosphorus compounds and amine compounds. In the case that an organic ~~phosphorous~~phosphorus compound is used, the one having a P=O linkage is an example. Such a compound is exemplified by phosphates such as trimethylphosphate, tri-2-propylphosphate, tributylphosphate, tetraisopropylethylenephosphonate, and phosphineoxides such as tributylphosphineoxide, trioctylphosphineoxide, and triphenylphosphineoxide.

Here, a water-extracting reaction in the case of using trimethylphosphate is expressed as the following formula (4).



It is expected that completely removing water is difficult even in the case that an extracting agent is added to a nonaqueous electrolyte solution as described above. Therefore, it is preferable to add, besides a water-extracting agent, a hydrofluoric acid-extracting agent which directly ~~remove~~removes HF to prevent metallic material from being corroded by HF. In addition, by adding a hydrofluoric acid-extracting agent alone to a nonaqueous electrolyte solution instead of a water-extracting agent, a hydrofluoric acid-extracting agent contributes to suppression of corrosion or the like of metal by HF, and thereby improvement in cycle characteristics is planned.

From such a ~~view point~~viewpoint, a hydrofluoric acid-extracting agent is suitably added to a nonaqueous electrolyte solution. Though a hydrofluoric acid-extracting agent can be used together with a water-extracting agent, it was found that a hydrofluoric acid-extracting agent greatly contributes to improving cycle characteristics even in the case that it is independently used as shown in results of the test described below as well as the case that a water-extracting agent is independently used.

Please replace the paragraph on page 40, line 22 – page 41, line 3, with the following amended paragraph:

As described above in detail, a lithium secondary battery of the present invention employs a nonaqueous electrolyte solution ~~where~~ in which is dissolved a lithium compound generating a lithium ion (Li^+) upon being dissolved as an electrolyte. Therefore, ~~there the invention is by no means limited with respect to the other material materials or a structure structures of the battery.~~ The main members constituting the battery and the structure are briefly described hereinbelow.

Please replace the paragraphs on page 41, line 22 – page 42, line 17, with the following amended paragraphs:

Another structure of an electrode body is a lamination type where a plurality of single-cell type of electrode bodies form a lamination. As shown in Fig. 2, a lamination type of electrode body 7 is formed by piling up positive electrodes 8 and negative electrodes 9 alternately via a separator 10, and at least one electrode lead 11 · 12 is attached to ~~one each~~ electrode 8 · 9. Materials for the electrodes 8 · 9 and methods for manufacturing the electrodes 8 · 9 are the same as the electrodes 2 · 3 in the wound-type electrode body 1.

Next, the structure is described in more detail with the example of the wound type of electrode body 1. The positive electrode 2 is produced by applying positive active material on both surfaces of a current collector. As a current collector, a metallic foil having good corrosion resistance against a positive electrochemical reaction is used, such as an aluminum foil and a titanium foil. Punching metal or mesh (a net) may be employed other than a foil. In addition, as positive active material, a lithium transition metal composite ~~oxides~~ oxide (e.g., LiMn_2O_4 , LiCoO_2 , or LiNiO_2) may be suitably used; and carbon fine powder of acetylene black or the like is preferably added thereto as a conducting aid.

Please replace the paragraphs on page 44, line 5 – page 45, line 24, with the following amended paragraphs:

The negative electrode 3 can be produced in the same manner as in the positive electrode 2. As a current collector of the negative electrode 3, a metallic foil having good corrosion resistance against a positive electrochemical reaction, such as a copper foil and a nickel foil is suitably used. As the negative active material, an amorphous carbon material such as soft carbon and hard carbon or a highly graphitized carbon powder such as artificial graphite and natural graphite can be used.

As the separator 4, there is preferably used the one having a three-layered structure where a Li^+ -~~permiable~~ permeable polyethylene film (PE film) having micro-pores is put between porous Li^+ -~~permiable~~ permeable polypropylene films (PP films). This doubles as a safety mechanism of controlling Li^+ movement, i.e., battery reaction in such a manner that, when temperature of the electrode body is raised, the PE film is softened at about 130°C to collapse micro-pores. Since the PE film is put between the PP films having higher softening temperature, PP films keep the shape and prevent the positive electrode 2 and the negative

electrode 3 from ~~a contact contacting~~ and producing a short circuit, and thus secure ~~suppress~~ suppression of battery reaction and security of safety become possible.

Upon winding operation of the electrodes 2, 3 and the separator 4, the leads 5 · 6 are attached to the electrodes 2, 3, respectively, in a portion where the electrode active material is not applied to expose the current collector. As the electrode leads 5, 6, the ones having a foil-like shape of the same material as the current collector of the electrodes 2, 3, respectively are shown. The electrode leads 5, 6 can be fixed to the electrode 2, 3 by the use of ultrasonic-wave welding, spot welding, or the like. At this time, it is preferable to fix each of the electrode ~~lead~~ leads 5, 6 so that an electrode lead of one of the ~~electrode~~ electrodes is disposed on an end surface of the electrode body 1 because the electrode leads 5, 6 can be prevented from ~~the contact with~~ contacting each other.

In composition of the battery, a produced electrode body 1 is inserted into a battery case with securing conduction between a terminal for taking out current outside and the electrode leads 5, 6 in the first place so as to be held in a stable position. After that, the battery is impregnated with a nonaqueous an-electrolyte solution; and then, the battery case is sealed to obtain a battery.

Next, a nonaqueous electrolyte solution to be used for a lithium secondary battery of the present invention is described. As a solvent, a single solvent or a mixed solvent of carbonates such as ethylene carbonate (EC), diethyl carbonate (DEC), dimethyl carbonate (DMC), propylene carbonate (PC), or γ -butyrolactone, tetrahydrofuran, acetonitrile, etc. can be used.

Please replace the paragraph on page 48, line 23 – page 49, line 13, with the following amended paragraph:

In Examples 1 – 3 and Comparative Example 1, there was found no difference in the surface form in the positive electrodes. However, as shown in Figs. 4(a) and 4(b), a large difference was observed in the negative electrodes. In Example 1, where the compound was added, a coat (lithium SEI layer) due to decomposition or the like of the electrode was observed on a carbon surface of the negative electrode as shown in Fig. 4(a). However, no other difference from unused carbon was found. On the other hand, in Comparative Example 1, a granulated substance was observed besides a lithium SEI layer on a carbon surface of the negative electrode as shown in Fig. 4(b). These negative electrodes were subjected to an EDS element analysis, and Cu was detected on the carbon surface and in the periphery of the carbon surface including the granulated substance. However, in negative electrodes in Examples 1 – 3, no Cu was detected.

Please replace the paragraphs on page 50, lines 10-23, with the following amended paragraphs:

As is clear from Fig. 5, a battery in Example 4 of the present invention achieved a capacity-retention rate of 93% in a 100-cycle test and exhibited by far excellent cycle characteristics in comparison with the one in Comparative Example 2, where the compound was not used. Thus, it has been clearly proved that a compound disclosed in the present invention exhibits excellent effect in a cycle life span, which is an important battery property, by an inspection in ~~the~~ Example 4, where water was intentionally added.

(Example 5)

A battery of ~~the~~ Example 5 was produced in such a manner that a coin-cell type electrode body was produced in the same manner as in Example 1 and put in a battery case, which was then filled with nonaqueous electrolyte solution.

Please replace the paragraph on page 56, lines 2-12, with the following amended paragraph:

As is clear from Fig. 8, batteries of Examples 7 – 11 achieved a capacity-retention rate of 82% in a 20000-cycle test and exhibited ~~by far~~ excellent cycle characteristics in comparison with Comparative Example 3, where the compound was not used. This seems that the compound containing an electron-donating element and a substituent inactivated HF in the electrolyte solution, and a salt of strong acid formed by a reaction of anion of inorganic acid in the compound and Li^+ covered a surface of the negative active material to suppress further formation of SEI, and as a result the cycle life span was improved.

Please replace the paragraph on page 57, line 14 – page 58, line 2, with the following amended paragraph:

As is clear from Fig. 9, batteries in Examples 12 - 14 of the present invention achieved a capacity-retention rate of 85% in a 100-cycle test and exhibited ~~by far~~ excellent cycle characteristics in comparison with the one in Comparative Example 4, where the compound was not used. Thus, it has been clearly proved that a compound disclosed in the present invention exhibits excellent ~~effect~~ effects in a cycle life span, which is an important battery ~~characteristics~~ characteristic, by an inspection in the Example, where water was intentionally added.

Please replace the paragraph on page 63, lines 2-9, with the following amended paragraph:

As is clear from Fig. 14, a battery in Example 22 of the present invention achieved a capacity-retention rate of 82% in a 20000-cycle test and exhibited ~~by far~~ excellent cycle characteristics in comparison with the one in Comparative Example 9, where the compound was not used. This seems that the compound containing an atom showing Lewis basicity inactivated HF in the electrolyte solution to improve the cycle life span.

Please replace the paragraph on page 66, lines 1-11, with the following amended paragraph:

Here, batteries of Examples 24 - 32 and Comparative Examples 11 - 12 were produced in a similar manner to that in Examples 7 - 11. Here, as the nonaqueous electrolyte solution, there was used a solution prepared in such a manner that LiPF_6 as an electrolyte was dissolved in a mixed solvent of an equivolume of EC and DEC so as to give a solution having a concentration of 1 mol/liter. In addition, ~~“NMP”~~ “NMP” in Table 10 means N - methyl - 2 - pyrrolidone, which is a solvent for dissolving the nonionic surfactant. All these various kinds of batteries had a battery capacity of about 10Ah after the charge in the first cycle.

Please replace the paragraph on page 66, line 23 – page 67, line 8, with the following amended paragraph:

Fig. 17 is a graph showing cycle characteristics of batteries produced with various nonaqueous electrolyte solution shown in Table 11. LiPF_6 was used as the electrolyte, and a mixed solvent of EC and DEC of the same volume was used as the organic solvent. These are common to all the samples. As shown in Table 11, triethylsilane was added as a water-

extracting agent in Example 33, tributylphosphate was added as a hydrofluoric acid-extracting agent, and both triethylsilane and tributylphosphate were added in Example 35. However, neither a water-extracting agent nor a hydrofluoric ~~acid-extracting~~ acid-extracting agent was added in Comparative Example 13.

Please replace the paragraph on page 68, lines 8-16, with the following amended paragraph:

Batteries of Examples 1 – 35 and Comparative Examples 1 – 13 were produced by the use of various battery-constituting members prepared by impregnating the inside of each battery case with the compound in the aforementioned method. In addition, the other members and environment for the test were made the same among all the samples, the battery members were sufficiently dried until the time just before assembly of each battery, and influence of penetration of water from outside of each battery due to insufficient sealing of the battery, or the like, was eliminated.